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Extraction of single bunches of synchrotron radiation from storage rings with an X-ray chopper based on a rotating mirror

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An ultrafast shutter has been developed for alteration of the time structure of synchrotron radiation from storage rings in the hard X-ray regime. In test applications on the wiggler beamline BW6 at DORIS, single bunches were extracted from the incident pulsed synchrotron radiation with minimum bunch-to-bunch distances of 482 ns. Even substantially shorter time windows may be defined in the case of tight collimation in the incident beam, *e.g.* on low-emittance sources. The shutter system is based on a new chopper concept involving a rotating X-ray mirror which totally reflects the incident radiation onto the sample through a remote slit. Rather low rotational velocities are sufficient to reach extremely short full open times. An additional shutter consisting of a slowly rotating disk prevents frame overlap and controls the repetition rate. A coincidence timing circuit checks the synchronization with the synchrotron bunch clock and provides trigger signals, *e.g.* for external excitation of a sample. The chopper system may be used, for example, in nanosecond time-resolved Laue diffraction experiments.

Keywords: time-resolved crystallography; Laue diffraction; stroboscopic data collection; nanosecond time resolution; X-ray chopper; mirror chopper; ultrafast shutter; alteration of the time structure.

1. Introduction

Crystal structure analysis on extremely short time scales below ~1 µs is of considerable potential interest for studies of structural intermediates. Considering small-molecule structures, one example is the possible study of electronically excited states with nanosecond lifetimes (Pruss, 1983; Bartunik, 1984). Materials science opportunities have been discussed by Larson & Tischler (1997). The feasibility of studies of protein structure and dynamics in the nanosecond to microsecond time range has been investigated by a number of authors (Bartunik, 1983; Moffat *et al.*, 1984; Moffat, 1989; Helliwell, 1992; Cruickshank *et al.*, 1992; Hajdu & Andersson, 1993; Šrajer *et al.*, 1996).

The pulsed nature of synchrotron radiation may be used for stroboscopic diffraction studies in the hard X-ray regime with an intrinsic limit in the time resolution of ~100 ps, as defined by the width of individual bunches at presently operating second- or third-generation storage rings. First uses of such a technique were reported for pulsed laser melting studies of silicon (Larson *et al.*, 1982, 1983, 1986) and for a study of a laser-pumped electronic excited state in cerium pentaphosphate (Pruss, 1983; Pruss *et al.*, 1984; Bartunik, 1984). In these applications the time course in scattering and diffraction was followed for a few reflections using single scintillation counters. For time-resolved three-dimensional structural analysis, a sufficiently complete set of structure-factor amplitudes is required. Reciprocal space may be explored with a minimum number of different crystal orientations by measuring a great number of simultaneously excited reflections with broad-bandpass Laue diffraction techniques. Methods of protein structure determination on the basis of Laue diffraction have been developed by a number of groups (Ren & Moffat, 1995; Campbell, 1995; Bourenkov *et al.*, 1996). Probabilistic procedures of Laue data evaluation, when applied with high-symmetry space groups, make it possible to derive three-dimensional structural information even from a single exposure (Bourenkov *et al.*, 1996).

Nanosecond to microsecond time resolution in X-ray scattering and diffraction experiments using synchrotron radiation may be achieved with a number of different techniques involving pulsed incident beams or gated area detectors (Larson & Tischler, 1997). In order to minimize the effects of radiation damage of protein crystals, the best solution is to define a time window in the incident X-ray beam containing a single bunch, and to repeat this procedure at a frequency depending on the excitation conditions until sufficient statistics are reached in the diffraction

Table 1
Bunch intervals τ_{BX} [µs]/electron current [mA] under different operating modes of the storage rings DORIS, ESRF and APS.

Mode	DORIS	ESRF	APS
Single-bunch	0.96/45	2.82/15	3.68/5
Two-bunch	0.48/85	–	–
Multi-bunch	0.19/140	1.88/200	0.18/100
Hybrid	–	0.94/7+193	1.23/5+95

pattern. Definition of a short time window may be achieved by deflecting the electron beam in the storage ring with a magnet (Nikitenko *et al.*, 1996). However, such a facility is not available on most storage rings, *e.g.* due to difficulties in inserting additional deflection magnets in existing lattices. An alternative solution is to block unwanted bunches with mechanical rotating or translating slits. A chopper system based on a rapidly rotating wheel for extracting a single bunch from the incident pulsed synchrotron radiation with 2.8 µs bunch intervals has been described previously (LeGrand *et al.*, 1989) and applied in a nanosecond time-resolved study of carbonmonoxy myoglobin following laser pulse photolysis (Šrajer *et al.*, 1996). Such rotating disk shutters require a high rotational speed; therefore, the technological difficulty and the costs increase rapidly when the bunch intervals are shorter. A newly developed phase-locked chopper with a basically similar concept has to rotate at supersonic speed (∼50 000 r.p.m.) in order to reach a full open time of 1.5 µs (Wulff, Bourgeois *et al.*, 1997; Wulff, Schotte *et al.*, 1997). On the other hand, the standard operating conditions of most synchrotrons involve even much shorter bunch intervals of <1 µs, since the vast majority of users are interested in high average flux or brilliance rather than time resolution and therefore request multi-bunch modes. A number of storage rings (Table 1) usually operate with bunch intervals between 180 and 190 ns (*e.g.* DORIS five-bunch mode, APS multi-bunch mode) and ∼950 ns (DORIS single-bunch mode; ESRF hybrid mode).

We solved the problem of extracting individual bunches under conditions of very short bunch-to-bunch distances by constructing a chopper system following a new concept. For the first time a rotating X-ray mirror is used as an ultrafast shutter. It deflects the incident pseudo-white beam onto a collimator slit near the sample position. Due to the narrow spatial angle under which the slit is seen from the mirror, the rotational velocities of such a mirror chopper may be reduced by more than one order of magnitude as compared with conventional solutions involving rotating disks. The present paper describes the concept of a rather inexpensive rotating-mirror chopper system that has been installed on the wiggler beamline BW6 at DORIS, and its performance in test applications. With the present version of the system, individual bunches may be extracted from the incident synchrotron radiation beam with bunch-to-bunch distances of ≥440 ns.

2. Instrumental design

The new chopper system consists of four main components (Figs. 1 and 2). Two of these components, including a rotating mirror (M) and a vertical slit (C3) at a distance R from M, form the ultrafast shutter part of the complete system and define the opening time. Two additional shutters, consisting of a slowly rotating disk (D) and an electromagnetic shutter (ES), prevent frame overlap and define the repetition rate. The complete chopper system is installed in a vacuum chamber.

The mirror M rotates around a horizontal axis which is perpendicular to the direction of the incident beam; this geometry takes the linear polarization of the synchrotron radiation in the orbit plane into account. The beam is reflected from M under a take-off angle $2\theta_M$ into the collimator. When M rotates, transmission through the exit slit C3 with vertical aperture A_{C3} is observed within a limited range, $\Delta\varphi$, in the rotation angle around $\varphi = \theta_M$,

$$\Delta\varphi \simeq (A_{C3} + 2W)/2R, \qquad (1)$$

where W is the vertical size (at FWHM) of the reflected beam that is incident on C3 at $\varphi = \theta_M$, and R is the distance between M and C3. The vertical aperture of the entrance slit, C2, of the collimator is assumed not to reduce the beam cross section. Rotation of M with a frequency ν_M defines the full open time, Δt,

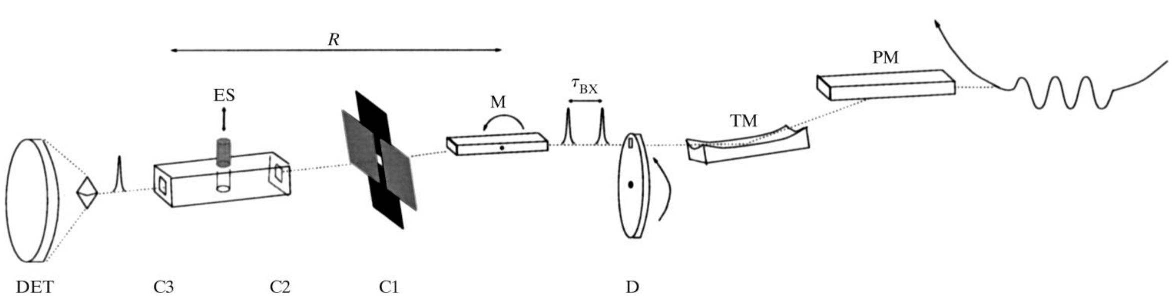

Figure 1
Scheme of the experimental set-up of the chopper system on BW6. The X-ray optics include a plane mirror (PM) and a toroidal mirror (TM). The chopper system includes a slowly rotating disk (D), a rotating mirror (M) and an electromagnetic shutter (ES). R is the distance between M and the collimator slit C3.

$$\Delta t \simeq [(A_{C3} + 2W)/2R]/(2\pi \nu_M). \quad (2)$$

The width of the selected time window decreases linearly with increasing rotational frequency. If the full open time is shorter than the time interval between subsequent bunches in the incident pulsed synchrotron radiation beam, the chopper system may extract a single bunch per period of the mirror rotation. If the rotating mirror is coated on either side, as is the case for the system installed on BW6, the maximum rate of transmitted single bunches is $2\nu_M$.

Depending on the excitation conditions for a given experiment, the repetition rate may have to be reduced, *e.g.* in order to avoid heat problems in the sample. The repetition frequency may be adjusted with the rotating disk shutter D which creates a time window Δt_D,

$$\Delta t_D \simeq [(s + h)/r]/(2\pi \nu_D), \quad (3)$$

where r is the radial distance of the slit aperture from the rotation axis of the disk, s is the horizontal slit aperture, and h is the horizontal width of the incident beam distribution. When ν_D is an integer fraction of ν_M, then D reduces the repetition frequency in the transmitted beam from $2\nu_M$ to ν_D. The number of repetitions may be controlled by a third shutter (ES) with opening and closing times of $<1/\nu_D$.

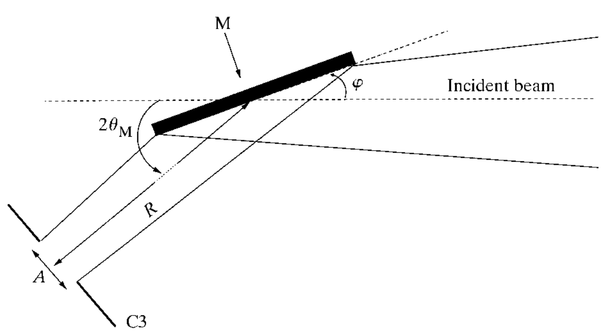

Figure 2
Geometrical conditions for transmission of the reflected beam through the vertical slit aperture of C3.

3. Motion control and triggering scheme

The rotating mirror (M) and the rotating disk (D) are driven by DC motors which are equipped with high-resolution incremental encoders. Both motors are controlled by an intelligent motion controller (IMC) which phase locks the rotation of the disk to the mirror rotation and provides trigger pulses for external use. The control software runs on a PC under Windows 95.

For each of the two axes, the IMC can generate a pulse of adjustable length at any specified phase angle of rotation. For D, the logic pulse is generated at an angular position where synchrotron radiation passes through its slit aperture. For M, the width of the pulse is chosen to correspond to the opening time Δt, and its phase is adjusted so that the pulse is generated one bunch interval before the mirror reaches its reflection position. These logical pulses may be used to set up a coincidence scheme (Fig. 3) for triggering, for example, a laser pulse for excitation of the sample. Assuming ES to be open and D in transmission position, a trigger is generated when the logical pulse for M coincides with a pulse of the bunch clock. The next bunch will hit the mirror when it is rotating through the acceptance angle $\Delta\varphi$ [equation (1)] for transmission. The trigger pulse is used to fire a laser at a time Δt_T before this bunch is transmitted and reaches the sample. In this way both excitation of the sample without irradiation with a synchrotron radiation bunch and the inverse situation are excluded.

The chopper system has been used in asynchronous mode. Phase shifts $> \Delta\varphi$ in the rotation angle cause a reduction in the duty cycle, hence an increase in the total time needed for accumulating a given number of synchrotron radiation bunches incident on the sample. Smaller phase shifts result in a reduction in the intensity in the transmitted bunch (see below). In both cases, however, the time resolution is not affected.

4. Performance

The chopper system which has been installed and tested on BW6 at DORIS consists of a rotating mirror, a slowly

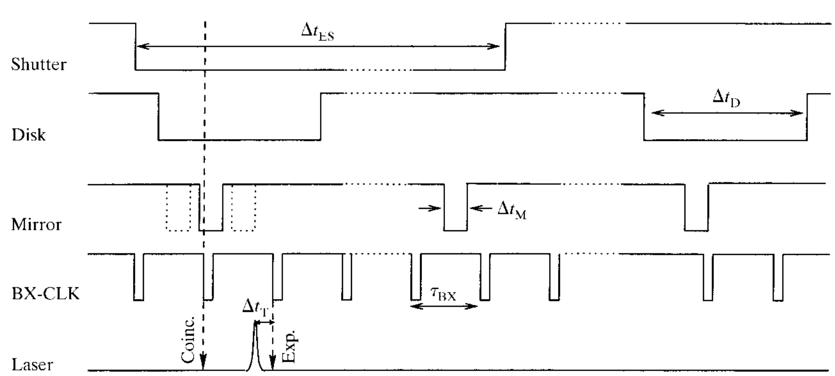

Figure 3
Coincidence circuit and trigger scheme. A trigger is created upon coincidence of four logical pulses. When the mirror is out of phase (dotted rectangles) the trigger conditions are not fulfilled.

rotating disk (r = 55 mm, s = 8.4 mm, h = 1.5 mm) and an electromagnetic shutter (opening/closing time ~20 ms). The complete system is operated in a vacuum. The rotating mirror of length 100 mm is made of Zerodur and has two gold-coated faces. The mirror reflects the X-ray beam under a take-off angle $2\theta_M$ = 3 mrad onto a collimator slit C3 at a distance R = 1340 mm. The X-ray beam which is incident on C3 has a vertical width of $W \simeq 0.33$ mm (FWHM). The vertical slit aperture is set to A_{C3} = 0.2 mm.

Using equation (2), the expected full open time may be estimated as a function of the rotational frequency ν_M (Fig. 4). A frequency ν_M > 53 Hz is required in order to create a time window of width $\Delta t < \tau_{BX}$ = 964 ns. τ_{BX} is the distance in time between two subsequent bunches at DORIS running in single-bunch mode. The transmission function has been predicted on the basis of ray-tracing calculations using *SHADOW* (Welnak *et al.*, 1994); the results are shown in Fig. 5. In these calculations the mirror was rotated in steps of 10 µrad through a total angular range of 0.22 mrad around the inclination angle θ_M = 2 mrad. For each step the number of rays passing through the collimator is displayed *versus* time, assuming that the mirror rotates with a frequency of 80 Hz. Vertical bars indicate the arrival times of individual bunches under conditions of pulsed incident radiation with 193 ns bunch intervals (DORIS five-bunch mode).

The theoretically predicted transmission function is in good agreement with the experimental results (Fig. 6). For M rotating at 80 Hz and DORIS running in five-bunch mode, in total four transmitted bunches were observed, in agreement with the theoretical prediction. The relative amplitudes of the experimentally observed bunches agreed within 10% with the predicted values (relative heights of the vertical bars in Fig. 5). The slight asymmetry that is visible in the tails of both the predicted and observed transmission functions arises from the angular distribution in the incident beam. A comparison between the experimental results (Fig. 6) obtained at rotational frequencies of 40 Hz and 80 Hz shows the linear dependence in the number of transmitted bunches on the mirror rotation period.

The length of the extracted bunch train was determined experimentally at four different rotational frequencies of M; DORIS was running in five-bunch mode. The results are shown in Fig. 4. The width of the time window was estimated from the number of transmitted bunches. The experimental tests were repeated when DORIS was running in two-bunch mode (bunch-to-bunch distance 482 ns). At a mirror rotation frequency of 80 Hz, corresponding to a time window with a full width of 580 ns, the chopper system extracted one single bunch (Fig. 7). In subsequent experiments we operated the chopper at even higher mirror rotation frequencies. At 105 Hz, using a slit aperture A_{C3} = 0.07 mm, a time window of 440 ns (full width at a 1% level) was defined. The maximum frequency that one can reach with the present encoder is ~145 Hz.

The rotational velocity of the mirror chopper is stable within a standard deviation of 8.7×10^{-6}. At a rotational frequency of 80 Hz, this corresponds to a phase shift or jitter of 109 ns in the location of the time window with respect to the time of arrival of a transmitted bunch. When the mirror chopper runs at a subharmonic of the bunch clock, a high duty cycle of ~80% may be reached even without phase locking, due to the high stability in the rotational movement. When operating the mirror at a frequency which did not correspond to a subharmonic, we obtained a duty cycle of 49%.

The width of the time window created by the chopper system in theory may be further reduced in the case of tighter collimation and smaller cross section in the incident X-ray beam. On low-emittance sources, for example, one may assume a small vertical size $W \simeq 0.1$ mm (FWHM) of the beam which is incident on C3. With a slit aperture A_{C3} = 0.1 mm at a distance R = 3000 mm and a rotational frequency ν_M = 100 Hz, the resulting full open time is ~80 ns. This estimate indicates that a mirror chopper, in combination with a slowly rotating disk D and a conventional shutter ES, may be used to extract a single bunch

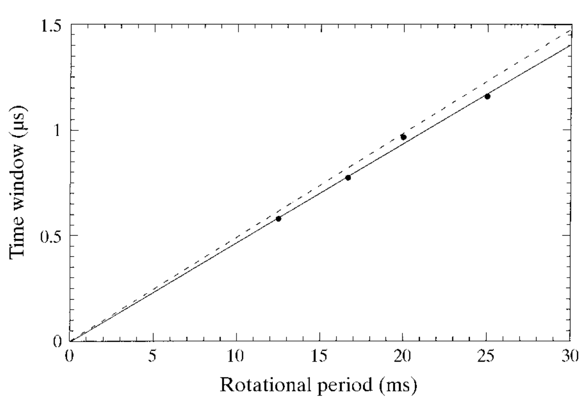

Figure 4
Theoretically predicted (- - -) and experimentally observed (●) full open times of the chopper system *versus* rotational period, t_M, of the mirror.

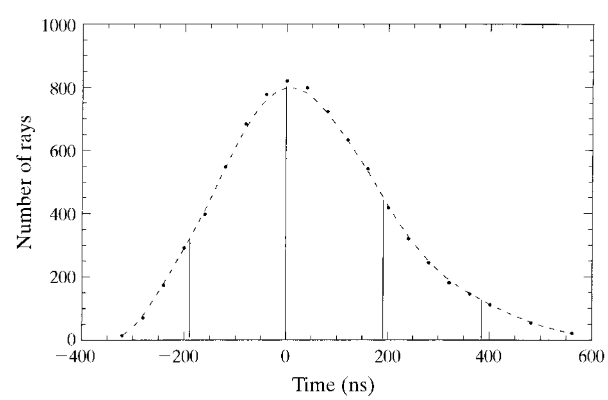

Figure 5
Transmission function obtained from ray-tracing calculations. The number of rays passing through C3 is plotted *versus* time.

from synchrotron radiation under multi-bunch conditions (Table 1).

5. Considerations for time-resolved Laue diffraction

For Laue diffraction experiments the mirror M (Fig. 2) has to totally reflect the incident X-rays over the entire wavelength and angular distributions in the incident X-ray beam when rotating through the angular range, ($\theta_M \pm \Delta\varphi/2$), corresponding to the transmission function of the chopper system. Hence, the glancing angle, ($\theta_M \pm \Delta\varphi/2$), of M must be smaller than the critical angle, θ_C, for the minimum wavelength present in the X-ray beam. The critical angle is (Batterman & Bilderback, 1991)

$$\theta_C \propto \lambda[\rho(Z+f')/A]^{1/2}, \tag{4}$$

where ρ is the density, Z is the atomic number, f' is the real dispersion term and A is the atomic weight. For a gold-coated mirror and X-ray wavelengths $\lambda > 0.55$ Å, as on BW6,

$$\theta_C\,[\text{mrad}] \simeq \lambda\,[\text{Å}] \times 7.1 > 3.9 \text{ mrad}. \tag{5}$$

θ_C is so far outside the range in the glancing angle that the transmitted bunch essentially will have the same spectral distribution as the incident pseudo-white beam. Taking the length of M and the divergence in the incident beam on BW6 into account, the maximum variation, at λ_{min} = 0.55 Å, in the reflectivity of the mirror is of the order of 1% during its rotation through an angular range $\Delta\varphi <$ 0.29 mrad (corresponding to $A_{C3} < 0.2$ mm) around θ_M = 1.5 mrad. A jitter in the rotational movement of M will not affect substantially the spectral distribution in the transmitted intensity. Therefore, accurate wavelength normalization is feasible; this is required for the evaluation of Laue diffraction patterns, in particular for deconvoluting energy overlaps by probabilistic methods (Bourenkov *et al.*, 1996).

The time resolution that can be achieved in the experiment depends on the width of an individual bunch (~100 ps) and the conditions of external excitation of the sample. Due to the coincidence circuit described above, a jitter in the mirror rotation has no influence on the time resolution.

6. Conclusions

The new chopper system permits extremely short full open times. This may be used to alter the pulsed time structure in the synchrotron radiation which is emitted from storage rings. The results of test applications of the system on the wiggler beamline BW6 demonstrate that a single bunch may be extracted from the incident synchrotron radiation from DORIS with bunch-to-bunch distances of 482 ns. The design concept of the new chopper system is based on the small angular aperture under which X-rays that are totally reflected from the rotating mirror pass through a remote collimator slit. Relatively small rotational velocities of the mirror are sufficient for defining much shorter full open times than with previously described ultrafast shutters based on rotating disks. The performance of the mirror chopper system may be easily further improved. On low-emittance sources the small cross section and tight collimation in the incident beam, on the one hand, and a longer mirror-to-slit distance, on the other, provide a basis for defining time windows of <100 ns. As a consequence, time-resolved X-ray scattering or diffraction experiments on time scales that are limited by the width of an individual bunch are feasible even under multi-bunch operation conditions.

In particular, the chopper system in combination with an integrating area detector is well suited for nanosecond time-resolved Laue diffraction studies of small-molecule or macromolecular structures. The rotating mirror totally reflects X-rays over a sufficiently wide wavelength range,

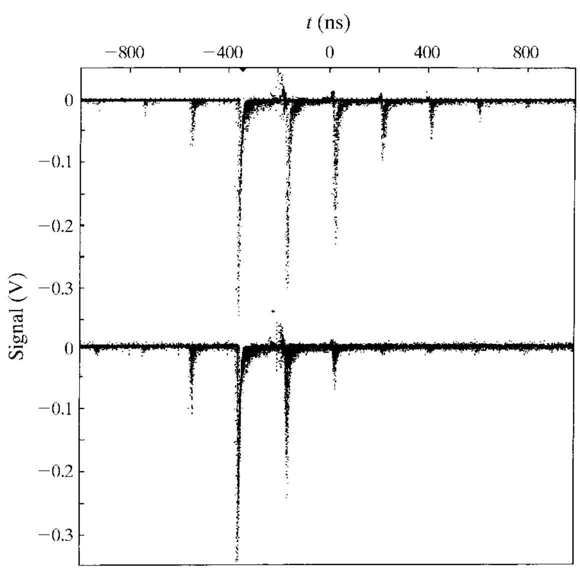

Figure 6
Oscilloscope recordings of X-ray pulses transmitted by the chopper system at mirror rotation frequencies of 40 Hz (top) and 80 Hz (bottom).

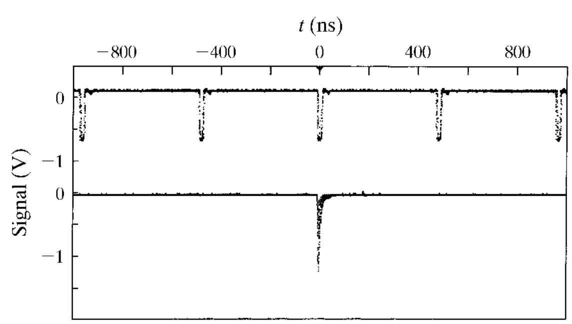

Figure 7
Single bunch (lower oscilloscope channel) which has been extracted from the incident synchrotron radiation beam from DORIS running in two-bunch mode. The upper channel shows the bunch clock signals.

and the rotational movement does not affect the wavelength normalization which is required for evaluation and scaling of structure-factor amplitudes. The repetition rate may be adjusted to the requirements of external excitation of the sample. A trigger is provided, for example, for firing a laser pulse at a defined time before the arrival of the synchrotron bunch to be transmitted. A coincidence circuit checks the relative timing of the chopper motions, the trigger and the bunch clock of the synchrotron. Thus, the transmitted X-radiation probes the sample under constant excitation conditions, independent of the number of repetitions.

The higher performance of the mirror chopper design as compared with previously described ultrafast shutters is achieved at rather low costs, since the considerable technological problems associated with the higher, by one order of magnitude, rotational velocities of rotating disk systems are avoided.

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